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Project Summary

Air Concentrations and Inhalation Exposure to Pesticides in the Agricultural Health Pilot Study

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The incidence of several types of cancers is higher among farmers than in the general population—this despite lower overall mortality. Occupational agents responsible for these excess cancers have not been definitively identified. The Agricultural Health Study seeks to identify and quantify pesticide exposures to farmers, indirect exposures to their families, and to assess health risks. A 6-farm, exposure pilot study implemented a total exposure assessment methodology, i.e., multimedia transport and multi-pathway exposure. Sampling design included air inhalation, oral ingestion, and dermal absorption. This paper reports on the air transport and inhalation exposures monitored during the exposure pilot study. Meteorological data were collected from an on-site three-meter tower. Outdoor air was sampled on the day of the pesticide application event, and indoor air samples were collected on three consecutive days centered on the application day. Personal activity logs, indicating time and location, were maintained by participants during the monitoring period. Of 33 targeted pesticides, 7 were applied on at least one of the participant farms, 11 were detected in the outdoor air near a farm residence, and 17 were detected in farm residence indoor air. Indoor concentrations of applied pesticides were detected on 4 of the 6 farms, however there is limited and conflicting evidence to support an exclusively outdoor air source of indoor concentrations of applied pesticides. Indoor concentrations of non-applied pesticides were more the rule than the exception. On 5 of the 6 pilot-study farms, concentrations of non-applied pesticides were detected in the indoor air sample on at least one day. As expected, the applicator's inhalation exposure to applied pesticides is greater than that of any other family member on the day of application. For spouse and children, the indoor microenvironment contributed to inhalation exposure of pesticides to a far greater extent than did the outdoor-on-farm microenvironment—even on the day of application.

This Project Summary was developed by EPA's National Exposure Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The incidence of several types of cancers is higher among farmers than in the general population—this despite lower overall mortality. Retrospective assessments of exposure to any suspected toxin are inadequate in determining environmental cause and health effect relationships. The Agricultural Health Study (AHS) is the first prospective study to evaluate the role of pesticides in cancer risks to farmers and their families. The AHS is a collaborative effort of the National Cancer Institute, the U.S. Environmental Protection Agency, and the National Institute of Environmental Health Sciences. The study seeks to identify and quantify pesticide exposures to farmers, and indirect exposures to their families, and to assess long-term health risks.

A total exposure assessment methodology was incorporated in the design of a 6-farm exposure pilot study, i.e., multimedia transport and multi-pathway exposure. Thirty-three pesticides were targeted. Sampling design included air inhalation, oral ingestion, and dermal absorption. Media and cohort monitoring were chronologically centered around farm pesticide application events. Baseline concentrations of pesticides were considered in sampling during a non-application (i.e., control) season, vs. the application season. This paper reports on the air transport and inhalation exposures monitored during the pilot study. Applicator, spouse, and up to two children participated from four Iowa and two North Carolina farms.

Air and Inhalation Exposure Monitoring Procedures

The assessment of direct inhalation exposure of the applicator during application events (handling, mixing, and loading (HML) operations; as well as actual pesticide application) required concurrent sampling of the applicator's breathing zone. Assessment of indirect inhalation exposures, as may be accrued by all family members from breathing indoor or outdoor air contaminated with fugitive pesticides, required sampling of indoor and outdoor air. All samples were collected with a polyurethane foam (PUF) and quartz pre-filter cartridge with a size-selective impactor at cartridge inlet which removed particles greater than 2.5 micrometers in diameter.

A five-day sampling strategy was chronologically centered on the day of a planned application event, hereafter synonymous with "day 3". The first and fifth days were directed toward setup and disassembly of monitoring equipment. During the second, or pre-application day, an indoor air sample was collected. During the application day, indoor and outdoor air samples were collected, as well as personal air samples from the applicator. During the fourth, or post-application day, an indoor air sample was collected. Participants' activity logs recorded the time, location, and activity of the applicator, spouse, and one or two children, during days 2, 3, and 4.

A personal air sampler measured the applicator's exposure to pesticides by inhalation during HML and application activities. A 3.8 L/min air sample was drawn through an inlet tubing positioned within the applicator's breathing zone. A meteorological monitoring tower collected wind speed, wind direction, temperature, and relative humidity data at 3 meters above ground level. Outdoor air pesticide concentrations sampled at the residences on the application day were expected to correlate with pesticide spray drift during application if meteorological conditions favored such transport. The indoor air samples, collected on days 2, 3, and 4, in conjunction with participant activity logs, provided data for indirect exposure assessment of all family members. A non-application season indoor air sample provided indication of baseline concentrations and chronic exposure. Indoor and outdoor samples were 24-hour averages; applicator personal air samples were collected over the duration of the activity of interest—either HML or application.

Modeling Initiative

The contribution of modeling in this study was to estimate potential peak outdoor concentrations under hypothetical near worst case conditions. The selection of application events suitable to a physically based model simulation was based on both model capability and data limitations. Each simulated case adheres to actual pesticide amounts applied and rate of application. However, meteorological conditions were a conservative composite of measured variables, and concentrations are calculated at plume centerline (i.e., assuming wind direction is directly from application field to monitor). Meteorological data are reported for selected farms concurrent with monitored application events.

The AgDRIFT model was initially developed to assess off-target drift deposition rates of water-based *aerial* pesticide applications. It can also calculate plume centerline *concentrations* needed in the assessment of inhalation exposure. At the model's core is a Lagrangian treatment of dispersion, tracking each nozzle stream of droplets through a flow field. AgDRIFT incorporates source constructs such as nozzle type, flow rates, and drop size distribution. Environmental variables having greatest impact on transport—wind speed, temperature, and relative humidity—are incorporated in AgDRIFT's calculations. AgDRIFT was deemed suitable to simulate *ground* boom sprayer drift, provided several extrinsic source parameters are appropriately assigned.

The application of the AgDRIFT model to ground boom spraying was accomplished by the appropriate setting of various emission and dispersion variables. The objective of the modeling exercise was to provide conservative assessments of downwind concentrations of applied pesticides for an averaging time typical of application duration. The actual application rate (pounds active ingredient applied per acre and per unit time) was adhered to in the calculations. Model simulations were consistent with measured values of wind speed during actual application periods. Quantitative details of pesticide application are reported, including pesticide identification, amount of active ingredient, total volume of liquid mixture applied, the concentration, acreage of application, application rate, duration of the application, and distance to the receptor (i.e., farm house). Additionally, the model estimate of worst case peak concentration at the receptor is reported, as is the "24-hour averaged" model calculation, and finally the measured 24-hour average concentration. Figures illustrate modeled peak one-hour concentration, at nominal adult breathing height (1.5 m), as a function of downwind distance.

Results and Discussion

Indoor and outdoor monitored concentrations of all detected pesticides are presented by farm, for both *control* (non-application) season, and application season. Indoor concentrations of an applied or residual pesticide were higher on the application day than conterminous days on Iowa farm #1 (dicamba), Iowa farm #2 (metolachlor), and Iowa farm #3 (alachlor; second application season). However, the cases observed in the pilot study do *not* strongly support a conclusion that outdoor air (exclusively) is the source of indoor concentrations of applied/residual pesticides.

The time, location, and activity of study participants were recorded by the participants during days 2, 3, and 4. These activity logs were reviewed with respect to participant location (and activity, in the case of HML or application). To assess exposure to detected pesticides, participant location was partitioned by characterized microenvironments (indoors; outdoors on farm; or performing HML/application activity), and time-in-microenvironment exposure was accumulated within 24-hour periods.

Applicator exposures represented the preponderance of the applied pesticide inhalation exposure of any family member on any AHS farm on the day of application. Applicators generally used no respiratory protection during HML or application activity. Not surprisingly, HML and application activities accounted for nearly all of the applicator's day 3 exposure to the applied pesticide. Applicator exposure from HML/application activity is presented as a percentage of day 3 total exposure. Applicator exposure during HML/application activities accounted for between 80% (alachlor on North Carolina farm #1) and 100%

(dicamba and 2,4-D butoxy ethyl ester on Iowa farm #2) of applicator day 3 exposure to applied pesticides. A summary of applicators' day 3 HML/application—related dose was also calculated with an estimated 25 liters per minute [L/min] breathing rate.

Inhalation exposures of all family members on the application day were calculated to assess relative exposure of applicator, spouse, and children. Application day air pathway exposures are presented by farm, applied pesticide, family member, and by microenvironment. The data are presented to permit attribution of a family member's exposure to each of the three designated microenvironments (i.e., HML/application activities, outdoors on farm, and indoors). When personal air samples distinguished multiple HML/application activities, separate exposures are calculated. The final column in each table provides the sum of exposures accrued in all microenvironments on the application day, by family member and applied pesticide.

The indoor microenvironment is unique in its contribution to pesticide exposure of *all* study participants via the air pathway. Time-activity logs indicated that all participating family members at all farms spent an average in excess of 8 hours per day indoors, over the three-day period. In all cases except Iowa farm #4, concentrations of at least one pesticide being applied, or found as residue within the application mixture, were measured indoors during the three-day monitoring period. Numerous examples of non-applied, non-ambient (no detectable outdoor concentration), pesticides were found to be present in indoor air samples. Indeed, indoor concentrations of non-applied pesticides were the rule more than the exception.

Indoor air exposures and inhalation doses of all indoor *detected* pesticides—*regardless of their known origin on the farm*, are presented for all participants, by farm, pesticide, and day. The three-day exposure sum, as well as a mean daily indoor inhalation dose, is also calculated in the final columns of each table. A resting breathing rate of 10 L/min was applied to exposure sums in calculating inhalation dose.

Summary and Conclusions

Of the 33 targeted pesticides, 7 were applied on at least one of the participant farms, 11 were detected in the outdoor air near a farm residence, and 17 were detected in farm residence indoor air.

The pesticide applicator was usually exposed to the applied pesticide(s) during HML and application activities. An exception was found on Iowa farm #4, where pyrethrins and piperonyl butoxide were applied but their presence in personal air samples was not detected. While the applicator's inhalation exposure to applied pesticides occurs almost exclusively during HML/application activities (at least 80%), exposure to non-applied (fugitive) pesticides does occur during time spent outdoors on the farm, and even more so during time spent indoors.

Outdoor concentrations of pesticides applied using a ground boom sprayer were detected (stationary point measurement) at significant concentrations (265 ng/m^3 alachlor, 24-hour average, on North Carolina farm #1) when wind direction favored such transport. The converse is not supported, however, as outdoor concentrations of atrazine were detected on Iowa farm #3 even though wind direction never favored source-to-receptor transport during HML or application activities. Outdoor concentrations of non-applied pesticides were detected on 5 of the 6 farms. The highest 24-hour outdoor concentration of any non-applied pesticide was 26.3 ng/m^3 metolachlor on Iowa farm #3 during the first application season, although metolachlor was found residual within the applied pesticide mixture. The highest 24-hour outdoor concentration of any non-applied, non-residual pesticide was 18.7 ng/m^3 alachlor, also detected on Iowa farm #3 during the first application season. Pesticide application events can substantially increase outdoor concentrations directly downwind—to levels exceeding typical applicator personal air concentration during handling, mixing, and loading. Elevated outdoor pesticide concentrations were not, however, clearly related to indoor concentrations on farms monitored in this study.

Indoor concentrations of applied pesticides were detected (stationary point measurement) on 4 of the 6 farms, although one case (metolachlor on Iowa farm #2) confirmed detection during the control season as well. Indoor concentrations of an applied or residual pesticide were higher on the application day than conterminous days on Iowa farm #1 (dicamba), Iowa farm #2 (metolachlor), and Iowa farm #3 (alachlor; second application season). However, the cases observed in the pilot study do not strongly support a conclusion that outdoor air (exclusively) is the source of indoor concentrations of applied/residual pesticides. Indoor concentrations of non-applied pesticides were more the rule than the exception. On 5 of the 6 pilot study farms, concentrations of non-applied pesticides were detected on at least one day. In most cases, applicator/spouse questionnaires relating to historical use of such pesticides could not confirm usage.

The applicator's inhalation exposure to applied pesticides is greater than that of any other family member on the day of application (calculated exposures indicated applicator inhalation exposure to be at least a factor of 5 times greater). This elevated exposure is attributable to HML/application activities. Non-applicator family members can be exposed indirectly to applied (and non-applied) pesticides during time spent indoors or outdoors on the farm. For spouse and children, the indoor microenvironment contributed to inhalation exposure of pesticides to a far greater extent than did the outdoor-on-farm microenvironment—even on the day of application. On two of six farms, mean daily indoor inhalation dose for a spouse or child was calculated to be on the order of micrograms—comparable to doses received by an applicator during an application (although toxicological response and health risk cannot be presumed to be the same). This importance of the indoor microenvironment to an individual's total inhalation exposure is attributable to several factors. Firstly, the time spent indoors (over the course of 24 hours) exceeded time spent outdoors on the farm. Chronic exposure to pesticides found in farm residence indoor air can be comparable, in cumulative inhaled dose, to exposures accrued by applicators during pesticide applications. Secondly, pesticide concentrations were generally higher indoors than outdoors (of 20 comparisons that could be made, 14 pesticides had higher 24-hour indoor concentrations; only 6 had higher 24-hour outdoor concentrations). Thirdly, the indoor microenvironment contained a greater number of detected pesticides than outdoors (17 different pesticides were detected in indoor samples; 11 were detected in outdoor samples).

Disclaimers

This paper has been reviewed in accordance with the United States Environmental Protection Agency's peer review and administrative review policies for approval for presentation and publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use. The information in this document has been funded by the National Institutes of Health and the United States Environmental Protection Agency. It has been subject to review by the National Cancer Institute and National Institute of Environmental Health Sciences and approved for publication.

*The EPA author, **John J. Streicher**, who is on assignment from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce, is also the EPA Project Officer (see below)*

The complete report, entitled "Air Concentrations and Inhalation Exposure to Pesticides in the Agricultural Health Pilot Study," (Order No. PB97-196 109; Cost: $25.00, subject to change) will be available only from:

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